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BACKWARD WAVE OSCILLATOR SOURCES FOR TERAHERTZ STUDIES

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ABSTRACT

In this report we will focus in particular on a new instrument that we developed with the BWO technology purchased with this grant. Briefly put, the BWO technology in this spectral region has ordinarily been operated at the end of a rather long, complex, slow, narrow banded, and expensive phase-locked frequency multiplication chain. We have demonstrated a new approach, FAst Scan Submillimeter Spectroscopy Technique (FASSST) which makes use of the very good short term spectral purity of the BWO sources and uses a fast scan, optical calibration, and fast signal digitization and computation to replace the phase-lock system. The result is a system of equal sensitivity and resolution, but one that is much simpler, less expensive, and orders of magnitude faster. In addition to numerous scientific and technical applications, the technique holds considerable promise for analytical chemistry and has been featured in an "A-pages" article in the American Chemical Society Journal *Analytical Chemistry*.

I. INTRODUCTION AND BACKGROUND

Although science and technology in the mm/submm spectral region have always been difficult, a number of important applications have emerged. Among these our laboratory has done significant work in basic chemical physics, quantum electronics, remote sensing, astrophysics, and atmospheric science. The BWO facilities developed have already made significant impacts on many of these projects. Brief descriptions of this work can serve to provide a flavor of this work:

(1) Basic Chemical Physics: In this work we seek to understand the dynamics of atomic and molecular collisions at very low temperatures. We are especially interested in these collisions because in the low temperature regime ($\sim 1 - 10$ K) the collision cross sections begin to vary rapidly with energy. Here the quantum mechanical nature of these collisions (which is washed out in the classical limit at ambient temperatures) dramatically reveals itself in the form of resonances which can be associated with quasi-bound states. This work is based on a new method which we have developed called collisional cooling which makes possible for the first time the study of equilibrium samples of gas phase molecules at temperatures far below their freezing points.¹⁻³ For technical reasons related to the relative sizes of $h\nu$ and kT , these studies are best carried out in the mm/submm spectral region.

(2) Quantum Electronics : Because of the technical difficulty of producing and detecting radiation in the mm/submm spectral region, we have long been interested in quantum electronic solutions to these problems and the scientific study of the phenomena which underlie them. For example, there are molecular lasers which operate in the mm/submm. Prior to our work, these were primarily discovered by trial and error and described by phenomenological models. Among other things, our detailed scientific studies of these systems revealed that fundamental limitations, which were believed to severely limit frequency tunability, were not in fact fundamental. Additionally, we were able to demonstrate that parameter spaces exist in which considerable tunability is possible.

(3) Laboratory Astrophysics: For decades the conventional wisdom was that molecular abundances in the interstellar medium were vanishingly small. This resulted both from an inability of theorist to postulate a formation mechanism and a recognition that the interstellar radiation environment would rapidly decompose the relatively fragile molecular bonds. However, by mm/submm radio astronomical techniques it was found that dense molecular clouds composed of mixtures of molecular gas and dust were abundant (the dust both shields the molecules from the radiation fields and are part of the solution of the formation problem). In addition to the intrinsic scientific interest in studying these clouds, these regions also are the regions of new star formation. As a result, this has now evolved into a major research area with a number of large, dedicated mm/submm telescopes and a number of major research groups. We have made many contributions to this field; perhaps the one with the most impact being the development and exploitation of a method of studying molecular ions in the laboratory which is the foundation of a number of major programs around the world.⁴

(4) Remote Sensing: In recent years there has been increased attention paid to the development of methods for the specific quantitative detection of trace species in the lower atmosphere. This is driven by everything from concerns about treaty compliance to the well known pollutant problems. Because the mm/submm region has a unique combination of capabilities for the penetration of particulates (clouds) and very high specificity and sensitivity, this application has been actively pursued even in the presence of the technical difficulties discussed above.

(5) Atmospheric Science: For many years it has been recognized that a complex cycle which involves both manmade and natural gases leads to the depletion of the ozone (O_3) layer. However, this complexity makes it very difficult to build, test, and verify the predictions of these models. As a practical matter, remote sensing techniques which observe the unique molecular spectroscopic signatures of the radiative emissions from these molecules in the upper atmosphere are used for this purpose, as well as for direct measurement of the ozone concentration. As an example of our work in this area, we have been involved with NASA in a project for the measurement of both the "dry air" and "moist air" continua, which make significant impact on both atmospheric remote sensing and propagation.

II. A DESCRIPTION OF THE FASSST SYSTEM

The FASSST system is based on broadband mm/submm BWOs produced by the ISTOK production company in Russia.^{5,6} Figure II-1 shows a block diagram of the system. In this example, an ISTOK OB-30 is used to cover the 240 - 375 GHz region. Similar tubes from ISTOK cover the ~100 - 1000 GHz region. The first wire grid polarizer (WG1) provides a well defined polarization from the output of the overmoded BWO waveguide. The second polarizer (WG2) is used to split the output power of the BWO, with ~90% being directed quasi-optically through the molecular absorption cell and detected by an InSb hot electron bolometer operating at 1.5 K. The remaining ~10% of the power is coupled into a Fabry-Perot (FP) cavity via a Mylar beamsplitter (BS1), which provides fringes for frequency interpolation between reference spectral lines of known frequency. In order to provide a highly accurate basis for the analysis of the frequency-voltage characteristic of the BWO, a folded FP cavity of length ~38.89 m is used to provide modes every ~3.854 MHz. A second molecular absorption cell that can be used for calibration purposes is also provided.

The key system elements include:

(1) The most fundamental element of the FASSST system is the excellent short term spectral

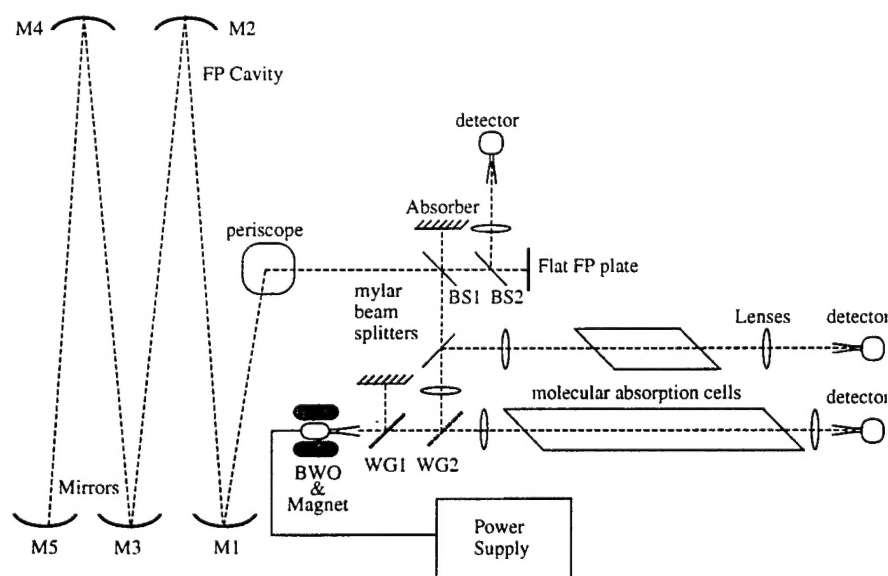


Figure II-1. Block diagram of the FASSST system.

purity of the BWO. From studies over many years, it has been observed that the short-term spectral purity of free running ISTOK BWOs is < 20 kHz.⁷ Without this spectral purity, the FASSST system would not be possible.

(2) Secondly, the BWOs can be voltage tuned continuously over an ~50% frequency range, which contains $\sim 10^5$ spectral resolution elements (Doppler limited).

(3) The synthesized

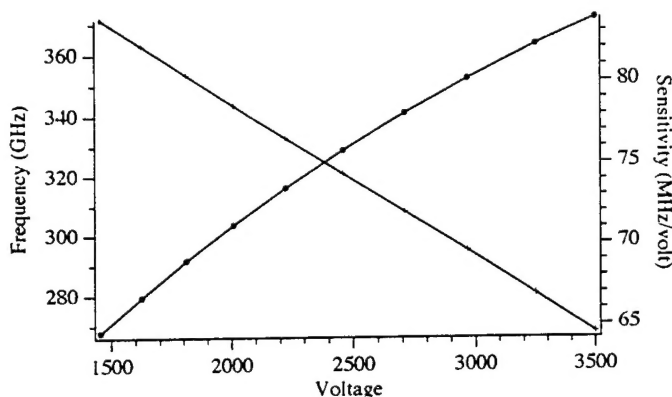


Figure II-2. Frequency - voltage characteristics of an OB - 30 BWO.

required to sweep from one reference fringe to the next. This eliminates the need for active frequency stabilization.

(5) Fast data acquisition and calibration hardware and software. In a very general sense, the bandwidth of this system plays the same role as the bandwidth of the lock loops of more traditional systems.

The combination of these five elements make it possible to measure thousands of spectral lines per second, with a frequency accuracy of a small fraction of a Doppler width ($\sim 0.1 \text{ MHz}/3 \times 10^6 \text{ cm}^{-1}$). Signal averaging is straightforward, and for equivalent integration times the sensitivity is the same as for slow-sweep, synthesized phase locked systems. Finally, the system is very simple in both concept and execution and can be used in a wide variety of applications.

Tube characteristics: The BWO is an electron beam device whose frequency depends on the interactions among the electrons, the periodicity of the BWO's slow wave structure, and the electromagnetic radiation propagating along with the e-beam in the slow wave structure. These are often described in the context of backward wave space harmonics and dispersion relations.⁸ Because the electron velocity has a first order effect in these relations, the frequency of oscillation f is strongly dependent on the electric potential V between the cathode and slow wave structure. For an OB-30, $df/dV \sim 75 \text{ MHz/volt}$. This increases somewhat with frequency and for tubes centered around 1 THz is of the order $\sim 100 \text{ MHz/volt}$. For a given tube, df/dV typically decreases with increasing voltage, largely because of the quadratic relation between electron energy and velocity. Figure II-2 shows the functional dependence of frequency as well as df/dV on voltage for the OB-30 used in this work.

Because the slow wave structure does not have a resonant frequency, BWOs can be very broadbanded. However, because of this low Q , feedback in the form of power reflected back into the slow wave structure can dramatically alter the frequency and power output of the BWO. In a broadband system in which fast, broad sweeps are executed it is not possible to control these reflections by narrow band matching techniques, and it is important that the reflections be carefully controlled and minimized. In fact, spontaneous locking to the FP etalon used as a frequency reference is not uncommon unless steps are taken to significantly reduce feed back.

The thermal drift of the frequency of the BWO can be $\sim 500 \text{ MHz/K}$ due to the thermal expansion of the slow wave structure. However, because the time constant for this drift is of the order of a few seconds (a time long in comparison to the interval between FP modes in the fast scan) it is not necessary to thermally stabilize the tube. This is fortunate because the power input to

frequency reference system typical of high resolution submillimeter spectrometers is replaced by a system more typical of optical spectroscopy. However, the longer wavelength significantly relaxes the requirements for optical precision and much greater frequency accuracy can be achieved.

(4) A fast ($\sim 10^5$ spectral resolution elements/sec, currently limited by detector bandwidth) sweep and data acquisition system 'freeze' any drift in the source frequency over the time

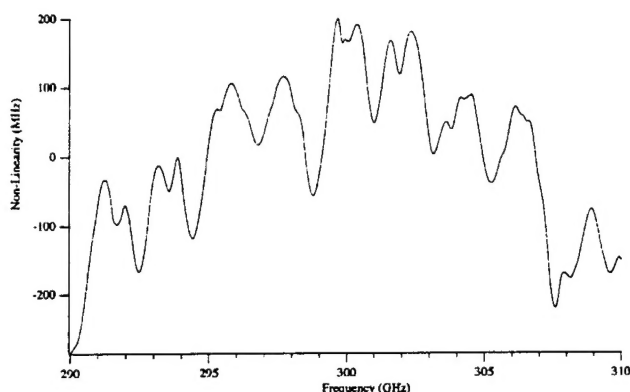


Figure II-3. Small scale fluctuations in the frequency - voltage characteristics of an OB - 30 BWO.

the BWO varies by more than a factor of two during the voltage sweep and external thermal control would be of little utility.

The frequency calibration scheme: If the frequency of the BWO as a function of voltage were linear, the frequency calibration scheme could be as simple as having two spectral lines of known frequency somewhere near the opposite ends of the frequency sweep range and using linear interpolation. However, from Fig. II-3 it is immediately obvious that the small scale structure is much more complex than given by a simple dispersion relation.

More specifically, if the effects of the small scale structure are not properly treated, the frequency accuracy of the FASSST system will be ~ 100 MHz, approximately 1000 times worse than that required for high resolution spectroscopy.

Based on these considerations, the basic FASSST scheme is to:

- (1) take a fast ($10^4 - 10^5$ MHz/sec) scan over the spectral region of interest,
- (2) include two or more (typically ~ 50 are available) reference lines,
- (3) use the known frequencies of the reference lines to determine the FP cavity mode spacing and absolute frequency,
- (4) count FP modes to establish the frequency of each fringe, and
- (5) use linear interpolation between the two nearest FP modes to calculate the frequencies of the unknown lines.

Because the thermal history of the BWO effects the frequency-voltage function at this level of precision, each sweep is calibrated separately.

The power supply scheme: Ultimately the combination of high spectral purity and voltage tunability of these BWOs makes the FASSST system possible. However, these qualities can only be exploited in the context of a power supply with complimentary properties, a stringent requirement. More specifically, the 240 - 375 GHz OB-30 BWO tunes ~ 100 GHz with a voltage variation of $\sim 1000 - 3500$ V, a tuning rate of ~ 75 MHz/volt. If it is desired that the voltage fluctuations of the power supply result in a frequency variation which is no more than 10% of a linewidth (~ 0.1 MHz), they must be less than ~ 2 mV in the context of a power supply capable of sweeping several thousand volts in ~ 1 millisecond.

The stabilization scheme: For the optical calibration scheme to be accurate, the BWO frequency drift must be linear over the time required to scan from one FP cavity mode to the next. In addition to thermal drift, power supply ripple can contribute to a non-linear sweep between FP cavity modes. In addition to fundamental power supply variations, voltages induced onto the beam accelerating voltage from the AC filaments of the BWOs will modulate the frequency of the system.

Resultant scan strategies: In our system the upper limit on the scan speed is set by the ~ 1 MHz bandwidth of the InSb detector. If the entire 100 GHz of the tube is to be scanned, Doppler widths are ~ 1 MHz, and 10 digitization points per spectral resolution width are required, the minimum sweep time set by the detector bandwidth is 1 second. This is a relatively long time, and the bandwidths required in the power supply sweeper would be low and reasonably straightforward to obtain. However, in many applications only a small portion of the 100 GHz available will be scanned, but the same sweep rate in terms of FP cavity modes/second is required in order to eliminate drift and ripple effects. As an example, if 20 FP cavity modes (~ 100 MHz) are scanned rather than the 20 000 in the full 100 GHz, the 1 second sweep period will be reduced to 0.001 second.

In both of these examples, the FP marker rate is 20 000/sec. If the ac ripple effects are of frequency ~ 100 Hz and magnitude V_{ac} , their maximum (depending on phase) contribution to the voltage change between FP markers will be reduced from V_{ac} by the ratio of marker rate to the ripple rate (a factor of 200 in these examples). Since only the nonlinearity of the ripple (which is small over $1/200$ of a cycle) contribute to calibration error, these fast scans effectively freeze ripple

effects. However, because the contribution of these effects to calibration error will grow as the inverse square of the scan rate, at slower scanning rates they can become significant.

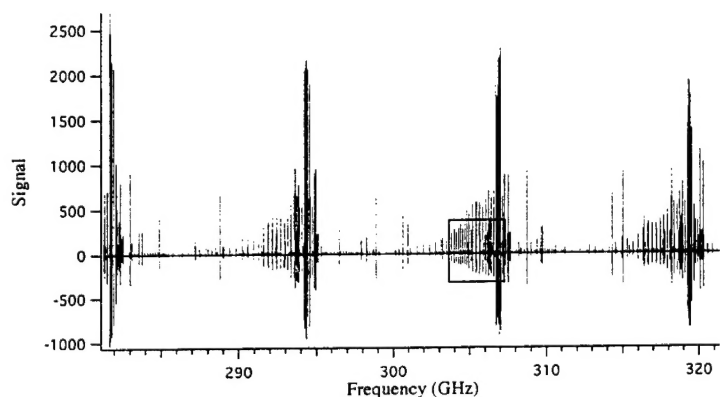


Figure II-4. An ~ 5 sec recording of 40 GHz of the nitric acid spectrum.

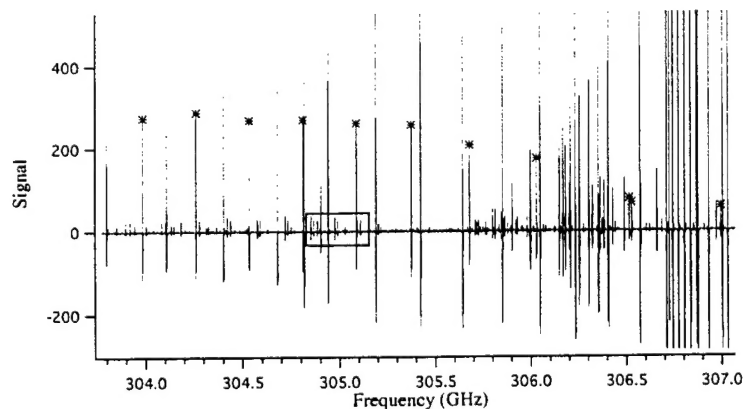


Figure II-5. A blow up of the box in Fig. II-4.

III. SOME REPRESENTATIVE RESULTS

Typical spectra: Because a single scan contains $\sim 10^6$ resolution elements, it is not possible to graphically display a complete, full band spectrum. However, Figs. II-4 - II-6 show a series of blow ups in both frequency and sensitivity of the mm/submm spectrum of nitric acid (HNO_3) which provide a good perspective. In the current system (which is not optimized for sensitivity), strongly absorbing lines are of $S/N = 10^4/1$ with a 10^{-6} sec integration time. Comparison of the closely spaced triplets in Fig. II-6 (which are due to the torsional motion in the v_9 excited state) with observations

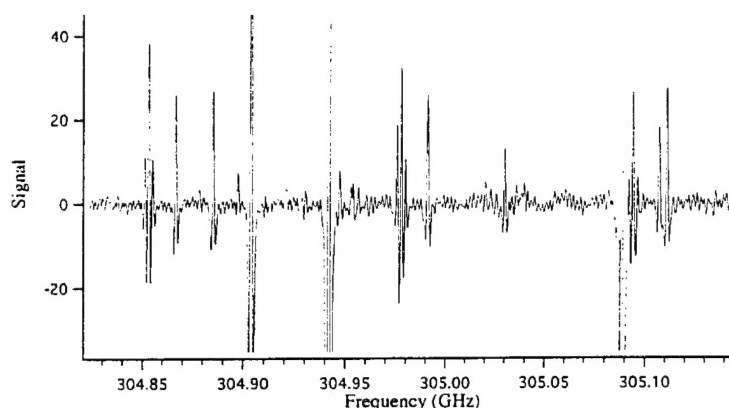


Figure II-6. A blow up of the box in Fig. II-5

of phase locked systems shows that the spectral linewidths recorded by the FASSST system are also Doppler limited.

Software details and operational practice: In order to efficiently utilize the FASSST system's ability to rapidly acquire and store spectral information, it is necessary to develop appropriate software and operational practices. It is

useful to first explore the information content of the spectra produced by the FASSST system. In $\sim 1 \text{ sec}$ 10^6 data points which characterize 10^5 resolution spectral elements can be acquired with a dynamic range of $\sim 10^4$. This is $\sim 10 \text{ MBytes/sec}$ of relatively uncorrelated data. Furthermore, because of the complexity and density of rotational spectra, many of these resolution elements may be filled with observable, resolvable absorptions. Although averaging multiple sweeps to increase S/N is possible, the inherent source brightness, detector sensitivity, and strength of the molecular interaction render this unnecessary in most applications. In these circumstances, real system efficiency is then limited by overhead: the cycling of samples through the system and more importantly by the time required to turn raw spectral information into useful scientific results.

In practice the operational procedures are:

- (1) The sample cell is filled to approximately 10 mTorr with the gas of spectroscopic interest.
- (2) The power supply initiates a sweep and triggers the computer to begin data acquisition.

Typically four channels are recorded: the output of the mm/submm detector in the absorption cell path, the output of the mm/submm detector of the FP cavity, the output voltage sweep, and the trigger. A full tube sweep ($\sim 100 \text{ GHz}$) takes 1 - 10 seconds and small spectral regions proportionately less time.

(3) If higher signal to noise is desired, additional sweeps are recorded. Since each sweep can be individually calibrated, no assumptions about tube or FP stability are required. Because mm/submm spectra typically contain baseline interference patterns in addition to the spectral information, provision is made for rapid gas fill/gas pump out cycles so that baseline subtraction is possible.

(4) The spectral and FP data file is then scanned automatically for peaks and fitting procedures used to locate (in bin or time space) each peak.

(5) For frequency calibration of the FP modes, the known frequency-voltage characteristic of the BWO tube is used to provide a first approximation so that Fourier transform correlation techniques between an observed reference spectrum and a spectrum calculated from the known spectral parameters of the reference gas can be carried out. In principle only two previously measured lines per calibration segment (which might vary between 1 and 100 GHz in width) are required to determine the fringe spacing and absolute frequency. However, in practice many known lines are used in a least squares procedure to determine these two parameters. While in principle the reference gas might be one selected especially for calibration purposes and contained

in the separate absorption cell, in practice many gasses have enough measured spectral lines to serve as their own self-reference.

(6) Linear interpolation is then used between each of the FP fringes to establish the frequency of each bin in the data. By comparison of the peak locations (which can be fractional bins) from step (4), the frequency of each unknown line is calculated.

(7) Because the absorption coefficients of spectral lines in the mm/submm can be calculated with good accuracy (often better than 1%), the known absorption coefficients of the lines of the reference gas are used to calibrate the absorption coefficients of the unknown lines, with linear interpolation used between the known lines. Over most of the BWO range, this absolute absorption calibration is good to ~10%. However, there exist narrow regions separated by ~10 GHz where the frequency and amplitude variations of the BWO are much steeper functions of the voltage. In these regions uncertainties of $\geq 100\%$ are possible. Clearly, in all regions the uncertainty in the absolute coefficient is directly related to the distance in frequency to the nearest calibration point.

Additional results: These facilities have been used to support a number of projects, including: Spectroscopic studies of Nitric Acid⁹, Dimethyl Ether¹⁰, Ethylene Oxide¹¹, Hydrogen Peroxide¹², Ethyl Alcohol¹³, Methyl Formate¹⁴, the development of Femtosecond Demodulation as a high spectral purity source for the mm/submm¹⁵, and the development of a new tunable mode-lock Ti:Sapphire laser system.¹⁶

IV. MAJOR PUBLICATIONS WHICH DESCRIBE THE FACILITY

"A fast scan submillimeter spectroscopic technique," D. T. Petkie, T. M. Goyette, R. P. A. Bettens, S. P. Belov, S. Albert, P. Helminger, and F. C. De Lucia," Rev. Sci Instrum. 68, 1675-1683 (1997).

"A New Gas-Phase Analytical Tool," S. A. Albert, D. T. Petkie, R. P. A. Bettens, S. P. Belov, and F. C. De Lucia, Analytical Chem. 70, 719A-727A (1998).

"Fast-scanning spectroscopic method for the submillimeter: the FASSST spectrometer," F. C. De Lucia and S. Albert, SPIE 3465, 236-246 (1998).